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PARAMETERS FOR ESTIMATING THE UPTAKE OF TRANSURANIC ELEMENTS BY TERRESTRIAL PLANTS

OFFICE OF RADIATION PROGRAMS

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U.S. ENVIRONMENTAL PROTECTION AGENCY
OFFICE OF RADIATION PROGRAMS
LAS VEGAS FACILITY
LAS VEGAS, NEVADA 89114

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16. ABSTRACT

This report summarizes information from the literature concerning parameters which can be used to estimate the transport of transuranic elements through plants to man. Plant uptake results from root uptake and deposition of contamination on above-ground surface areas of the plant. Deposition on plant surfaces occurs from both the initial contamination cloud and resuspension of contaminated soil. Generally, a deposition interceptor factor (pCi on vegetation per unit area subtended by the vegetation, per pCi per unit area of ground) of about 0.2 is indicated. A weathering half-life of about 30 days for time periods of about a week after deposition is suggested. Plant uptake parameters generally range from 10^{-6} to 10^{-3}; however, some data has been extrapolated to estimate a long-term uptake (20 years) of 0.3.

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PREFACE

The Office of Radiation Programs of the U.S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety. This literature survey was undertaken to assess the available information of parameters for estimating the uptake of transuranic elements by terrestrial plants. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

Donald W. Hendricks
Director, Office of
Radiation Programs, LVF

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INTRODUCTION

This report summarizes information from the literature concerning parameters which can be used to estimate the transport of transuranic elements through plants to man. The scope of the report is limited to parameters for estimating the concentrations of transuranics in terrestrial plants based on activity concentrations in soil and air.

There is only a limited amount of information specifically concerning plant uptake of transuranics. In many instances it has been necessary to use information based on other elements, which interjects additional uncertainties due to the variance in physical and chemical characteristics of these elements versus the transuranics. Furthermore, most of the transuranic data relates to plutonium; thus, this report focuses on plutonium.

Brown (1975) presents a bibliography of information concerning plant uptake of americium. Americium has only received cursory coverage in this review, although consideration has been given to the differences between americium and plutonium in plant uptake. Differences in the mobility and uptake of plutonium-238 and plutonium-239 are discussed. Papers concerning deposition and retention of plutonium on reindeer lichens have been excluded.

Plant uptake results from root uptake and deposition of contamination on above ground surface areas of the plant. Although deposition or fallout on the plant may not actually be taken into the plant tissue structure, Romney et al. (1975) and Hanson (1975) note that it may be tightly bonded to the plant microstructure and become essentially indistinguishable from material in the plant tissue. Deposition on plant surfaces occurs from both the initial contamination cloud and resuspension of contaminated soil.

SUMMARY AND CONCLUSIONS

The limited amount of available information is not adequate to select precise parameters for estimating concentrations of plutonium in vegetation due to root uptake and foliar deposition. The existing data base is essentially non-existant for some parameters, and shows significant variations for other parameters. Furthermore, much of the existing laboratory-generated data is not directly applicable to the normal geographical and climatological field conditions.

There is a significant variance in the estimates of the particle size distribution for airborne plutonium. Estimates of aerodynamic mean diameters appear to vary from sub-micrometer to about 10 μm diameter particles. For given particle size distributions, there are uncertainties in the deposition velocity, vegetation interception factors, and retention parameters. Furthermore, much of the experimental data appears to be for particle-size distributions significantly larger than those expected from normal nuclear reactor fuel cycle plant releases, worldwide fallout, or resuspension. There is the additional unknown feature in that specific plant deposition parameter work has not been done with plutonium.

The plant deposition parameter information is summarized in Table 1. These data imply a deposition interceptor factor (F, pCi on vegetation per unit area subtended by the vegetation, per pCi per unit area of ground) of about 0.2. Although the measurement of this parameter is time dependent, the time after deposition is generally not indicated in these studies. It appears that the weathering half-life is short (hours to a day) during the initial deposition period.

There is a significant range in the initial retention estimates associated with the type of vegetation, and more importantly, a variance associated with the time after deposition when the measurement is made. The intervening wind and precipitation conditions are also of prime importance.

It is suggested that the initial retention and weather half-life data should be treated as sets for each individual study. That is, the individual parameters should not be averaged between studies without a detailed evaluation as to common situations (e.g., time of measurements and climatology). Prudence appears to indicate choice of a weathering half-life of about 30 days for time periods of about a week after deposition. For small plants,

not prone to trapping particles, a retention factor of about 20 to 30 percent appears reasonable, but there is considerable uncertainty in the data.

Plant uptake parameters are summarized in Table 2. Hanson (1975) estimates the plutonium uptake by plants to be about 10^{-4}. Others generally categorize it as about 10^{-3} to 10^{-6} which includes most of the data in Table 2. Romney et al. (1970) and Neubold (1963) present data that show a definite increase in plant uptake with successive crops. Romney's et al. (1970) data indicate about an order of magnitude increase, from 1.9×10^{-5} to 14×10^{-5} over a 5-year period. This increase is generally related to microorganism activity in the soil (Au (1974), and Au and Beckert (1975)) and chelation by organics in the soil (Romney et al. (1970)).

There is some indication that plutonium-238 is more mobile than plutonium-239, but this has received only limited verification. Cline (1967) reported that barley took up 50 times as much americium-241 as plutonium-239. Romney et al. (1974 and 1975) have also reported that americium-241 appeared to be more mobile than plutonium.

Essentially all of the plutonium uptake studies are based on laboratory experiments containing plutonium uniformly mixed throughout the soil volume thereby increasing root contact. Results therefore appear to be unrealistic for natural vegetation where the deposited plutonium is largely limited to the upper 2 to 5 cm of soil, above the natural root mat. Therefore, the laboratory results should be conservative for most natural plant species growing in undisturbed or unplowed land; but, the uniform distribution of the plutonium in the laboratory soil should be representative of farm crops grown on plowed land.

Bloom et al, (1974) and Martin et al. (1974) report data from Romney et al. (1974) indicating plant uptake from Nevada Test Site field studies. A total plant uptake of about 0.3 is inferred. They also estimate a total long-term uptake (20 years) of about 0.3 by exponentially extrapolating Romney's et al. (1970) 5-year study to 20 years. Romney et al. (1974 and 1975) qualify the 0.3 uptake as being from both root uptake and deposition and they emphasize that deposition is the primary contributor, probably by several orders of magnitude.

There appears to be general consensus that deposited plutonium is not taken up (by foliar absorption) into the plant; rather, it is generally immobile. This hypothesis is based on limited information.

DEPOSITION ON PLANT SURFACES

The following parameters are used in the deposition pathway:

- Deposition velocity: This may be given with respect to the ground surface or for vegetation. If the deposition velocity for the ground is used, the plant intercept area or factor must be used.

- Plant interception factor (F): Witherspoon and Taylor (1969) defined $F=WC^{\circ}/m$,

  where:

  W is the biomass of foliage in grams (dry weight) per square foot of soil surface area (g/ft^2).

  C° is the quantity of radionuclide initially intercepted per gram dry weight of foliage ($\mu Ci/g$).

  m is the quantity of radionuclide deposited per ft^2 of soil surface area.

  Thus, F is the ratio of radioactivity deposited on the foliage to the radioactivity deposited on the ground area inhabited by the foliage. The ratio has no units. The product of the deposition velocity for the ground surface and F is the initial effective deposition velocity for vegetation.

- The initial retention factor, (f), is the fraction of radioactivity originally deposited that remains at some time after deposition. This is usually given for one to two weeks after deposition.

- The weathering or retention half-life ($T\omega$) represents the exponential decrease in the retention of deposited activity after the initial one to two week period.

- The plant biomass of foliage (W) is given as grams dry weight per square meter.

- The fraction of deposited activity that is actually taken into the plant is denoted as (d).

CONSIDERATION OF PARTICLE SIZE

The deposition velocity, plant interception factor, and plant retention of deposited material are all a function of the particle size distribution. The size distribution of airborne particulates is related to the source of release. Particle size distributions vary from the micrometer and sub-micrometer diameter for nuclear fuel cycle plants and worldwide fallout to tens of micrometers for near-in fallout and aged fallout in the soil.

High efficiency particulate air filters (HEPA) are used to minimize releases from nuclear fuel cycle plants. HEPA filters have removal efficiencies of 99.97 percent for 0.3 μm diameter dioctyl phthalate smoke particles; thus, releases from most nuclear installations are assumed to be in the sub-micrometer size range (Burchstad (1967)). Moss et al. (1961) also report mass median diameters of less than 1 μm for airborne plutonium in working areas of a plutonium fabrication plant.

Klement (1965) indicates that particulates from nuclear explosions are generally in the sub-micrometer range; but, they may become attached to other material forming conglomerates of 10 μm or more (Gudiksen and Lynch (1975), and Nevissi and Schell (1975)). Generally, worldwide fallout is classed in the micrometer to sub-micrometer diameter size.

The size distribution of resuspended material is dependent on both its original size and composition, and on the material to which it becomes conglomerated within the soil. Bretthauer et al. (1974) analyzed particles from air samples at the Nevada Test Site (NTS) and observed plutonium-bearing particles from less than 0.5 to 17 μm in diameter. The composition of the particles ranged from plutonium, uranium, and oxygen (several micrometer) to silicate and organic particles (about 10 μm). The geometric mean particle diameter was about 1.5 μm.

Volchok et al. (1972) report data from two studies of airborne particulates around the Rocky Flats Plant in Colorado. The initial study, based on particles on particulate filters, indicated a mean diameter around 10 μm. This study was potentially biased by the lack of analysis sensitivity for particles below 0.5 μm. Results from six cyclone and elutricator samples (run time of about 50 hours) indicated median diameters of about 5 μm.

Tamura (1974 and 1975) reports on the particle size distribution of plutonium in NTS soils. One to ten percent of the activity was found in the 0 to 5 μm diameter soil fraction; whereas, about 60 percent (up to 90 percent for several samples) of the activity was found in the less than 53 μm size. Romney et al. (1975) indicate that the micro-structure of many species of vegetation is adept at capturing particles of these size ranges. The bond between the vegetation and these particles is

apparently so tenacious as to make it almost impossible to distinguish the material from that actually taken up into the plant.

Bloom et al. (1974) assume a particle size of 10 μm for plutonium in air, which leads to a value of about 5 cm/sec for the deposition velocity. These values are used in their environmental plutonium model.

Bagnold (1945) and Chepil (1945a,b,c,d) indicate that over 90 percent of the wind movement of soil is by surface creep and saltation. These phenomena occur at heights below 1 meter above the ground surface and are not observed on standard air samples. Surface creep and saltation are connected with movement of soil particles of tens to hundreds of micrometers in diameter. Thus, they include movement of the soil size fraction that contains the majority of plutonium (Tamura (1974 and 1975)). Furthermore, vegetation can retain particles of this size class, (Romney et al. (1975)).

In summary, various investigators recommend a range of particle sizes for airborne plutonium. Particle material from original source terms is generally in the micrometer to submicrometer class. Plutonium in soil (limited data, mostly from NTS) appears to be predominately associated with particles between 20-50 μm in diameter (Tamura (1974 and 1975)). The limited information from Bretthauer et al. (1974) and Volchok et al. (1972) indicate that the mean diameter of resuspended plutonium particles is less than 10 μm, probably around 5 μm. Much of the activity deposition on plants with foliage near the ground, would appear to result from surface creep and saltation associated with the larger diameter particles (10 to 100 μm) versus resuspended material.

DEPOSITION PARAMETERS

Witherspoon and Taylor (1969) present data for simulated fallout on pine and oak trees, using cesium-134 as a tracer. The initial fraction (F) of the simulant fallout (88 to 175 μm diameter particles) intercepted and retained by foliage was higher in the oak tree (0.35) than in the pine tree (0.24). After 1 hour, the broad-leaved oak lost about 90.5 percent of the initial deposition, while the pine loss was only about 10 percent, corresponding to initial retention factors of 0.095 and 0.90, respectively. Weathering half-lives (T_ω) due to wind, rain, and all other environmental factors were determined to be 25 and 21 days for the oak and pine trees, respectively, for the period from 7 to 33 days after initial fallout deposition.

Witherspoon and Taylor (1970) present data for five crops using simulated fallout with rubidium-86 as a tracer. Two sizes of quartz particles (44 to 88 μm and 85 to 175 μm diameter) were used on squash, soybean, sorghum, lespedeza, and peanuts. For the size range 44 to 88 μm, the fraction of fallout initially intercepted (F) ranged from 0.075 for the small-leaved lespedeza to 1.248 for the squash. Interception factors (F) greater than unity were obtained for squash and soybean plants. Such plants, which have bush-like structures, have large exposed surface areas available in many different interception planes. The average fraction intercepted (F) for the smaller diameter particle size range was 0.587, which was about 2.5 times greater than F for the larger particle size range.

Fisher (1966) predicts a theoretical decrease in the deposition velocity on pasture grass with decreasing particle size in the 20 to 0.1 μm range. It would appear that using Witherspoon and Taylor's data for the 44 to 84 μm range would be conservative, but there is limited information on which to base this hypothesis.

Witherspoon and Taylor (1970) also studied particle retention. Losses from plant foliage due to wind removal (during the first 12 hours postdeposition) ranged from 3 to 35 percent (average--21.1 percent) for the 44 to 88 μm particles. During the same period, losses for the larger particle size ranged from 9.5 to 26 percent (average--15.8 percent). For the 12- to 36-hour period postdeposition, losses ranged from 1.2 to 33.5 percent (average--15.4 percent) for the smaller size simulant and ranged from 7.7 to 34 percent (average--21.6 percent) for the larger size simulant. Therefore, during the first 12 hours of postdeposition, when the wind speed averaged 0.5 mph and there was no rainfall, the plants lost an average of 18.5 percent of the initial deposition. This corresponds to an average value of the initial retention factor (f) of 0.815. Losses for the next 24 hours also averaged 18.5 percent. From 1.5 to 7 days postdeposition, the plant retention dropped from 63 percent to about 33 percent. This decrease was largely related to 0.25

inches of rain on the sixth day. Retention dropped to 7.9 percent of initial deposition during the second week, after a heavy rainfall of 1.33 inches. Another intense rainfall caused a sharp decrease in fallout retention to 3.3 percent, which slowly decreased during the remainder of the experiment.

The weathering half-lives ($T\omega$) for the 44-84 μm particles were 26 days for the 14- to 28-day postdeposition period and 84 days for the 28- to 56-day postdeposition period. During the entire 56-day study period, the weather half-lives ($T\omega$) ranged from 2.09 to 272.8 days for the 44 to 88 μm particle size; and 1.33 to 56.5 days for the 88 to 175 μm particle size. This experimental data indicates that large differences in initial interception existed between plant species for different particle size distributions, but that these differences become insignificant after about 1 week of exposure to environmental influences such as wind and rain.

Witherspoon and Taylor (1970) report values for the activity per dry gram of foliage, per activity per unit area of soil for the five crops. These values range from 0.01 to 0.2 ft^2/g (10 to 200 cm^2/g). The biomass values (W), in grams of dry foliage per square meter of soil (dried at 100°C for 24 hours), ranged from 20 for lespedeza to 120 for soybeans. These data are for 6-week-old plants at the time of deposition. The plants were planted the last of May in the Oak Ridge, Tennessee, area. All of the foregoing values are summarized in Table 1.

Concentrations of iodine-131 and strontium-89 on plants contaminated by fallout from Project Sedan at the Nevada Test Site were reported by Martin (1965). Examination of the fallout deposited on foliage indicated that most of the activity was due to particles less than 5 μm diameter, with virtually no retained particles greater than 44 μm diameter. The observed effective half-lives for iodine and strontium on the vegetation corresponded to weathering half-lives ($T\omega$) of 17 and 28 days.

Russell (1965) presents a review of interception and retention of airborne material by vegetation. Based on data from Milborn and Taylor (1965) concerning strontium-89, Russell concludes that on the average nearly one-quarter of the deposited fallout material is initially held on edible leaf tissues. An equal quantity may be associated with the basal tissues. The studies of Milbourn and Taylor also indicate that 50 percent of the radioactivity present on the edible herbage per unit area is usually lost in about 14 days. The fraction of initially deposited fission products lost from cabbage plants in a 28-day period ranged from 0.83 for cesium-137 to 0.90 for ruthenium-106 (Middleton and Squire (1961)). Data were also presented which indicated that washing cabbage leaves in water could remove from 10 to 36 percent of the deposited contamination (average of 24 percent). Middleton and Squire also concluded that the extent of radionuclide absorbtion into leaves was of little importance

TABLE 1. SUMMARY OF PLANT DEPOSITION AND RETENTION PARAMETERS

Reference	Plant Interception Factor F	Foliage Biomass (g,dry/m^2)	Initial Retention Factor	Weathering Half-Life (Days)	Half-Life Pertinent Period(Days)	Comments
Witherspoon & Taylor (1969)	0.35		0.095	25	7 to 33	88 to 175 μm particles, oak trees
Witherspoon & Taylor (1969)	0.24		0.90	21	7 to 33	88 to 175 μm particles, pine trees
Witherspoon & Taylor (1970)	0.075 to 1.2	20-120	<0.1@ 2 wk	26	14 to 28	44 to 88 μm particles, 6-wk old plants
Witherspoon & Taylor (1970)		20 lespedeza	~0.8@ 12 hr			
Witherspoon & Taylor (1970)		120 soybeans	~0.3@ 1 wk			
Levin et al. (1970)	0.25	540 fruits		14		
Levin et al. (1970)		3500 leaves				
Martin (1965)				17		Fallout, I-131
Martin (1965)				28		Fallout, Sr-89
Russell (1965)			~0.5			
Russell (1965)			~0.17			Cabbage, Cs-137
Russell (1965)			~0.10			Cabbage, Ru-106
Bloom et al. (1974)	5 cm^2/g			30		F factor divided by biomass.
Bloom et al. (1974)	~ 0.14					Assume biomass of 289g/m^2
Martin et al. (1974)	5 cm^2/g			30		
	~ 0.14					
Milbourn & Taylor (1965)				14		Project Sedan Sr-89

compared to the retention of activity on the leaves.

Levin et al. (1970) discuss the choice of parameters for dose model calculations for the proposed nuclear applications in the construction of an interoceanic canal. The information they present is oriented to South America and general fallout of mixed fission products. Thus, the information has limited applicability to conditions in the United States. A summary of their parameters is given in Appendix A. Several of the more pertinent values are:

- --Fraction of the element in plant edibles which comes from leaves (due to foliar deposition) of 0.05. This value apparently relates to fruit type plants.
- --W, biomass of plant edibles of 540 g/m^2 (dry weight). Biomass of plant leaves 3500 g/m^2.
- --Growth-rate coefficient for plant edibles of 0.05 day^{-1} or half-life of 14 days.
- --Weathering elimination rate for plant leaves of 0.05 day^{-1} or 14-day half-life.
- --F, Fraction of fallout intercepted by plant leaves of 0.25.

The weathering elimination rate ($\lambda\omega$) was estimated to be 0.05 day^{-1} based on a $T\omega$ of 14 days. The fraction of fallout intercepted by plant leaves was 0.25. For most fission products the fraction in the plant edibles which comes from leaf contamination was estimated to be 0.001. The fraction in plant edibles which comes from the root uptake of contaminants in soil was 1.0. These values are estimates influenced by natural weathering conditions and decontamination due to washing and food preparation.

Bloom et al. (1974) review the literature values to obtain parameters for use in their transport model. Based on fission product fallout data, they postulate the following half-lives:

Half-Life (Days)	Time Increment (Days)
1.4	0 - 5
20	5 -15
30	15 -30
130	30 -60

Bloom et al. (1974) note that such parameters as interception, retention, and retention half-life are dependent on the time after deposition when these factors are measured. Given this, they select an interception factor (units of cm^2/g dry weight) of 5 and a weathering half-life of 30 days. This interception factor (in units of cm^2/g) is equivalent to the unit-less interception factor (F) divided by the plant biomass

(W) in g/cm^2. Bloom et al. (1974) refer to the studies by Miller and Lee (1966) of interception factors (cm^2/g) for fallout from a volcano eruption. Values of the interception factor vary by a factor of two (from 47 to 96 cm^2/g) between low and high (greater than 90 percent) relative humidity. Miller and Lee (1966) note that these values were based on samples collected immediately after deposition. Notation of time after deposition may explain the discrepencies with nuclear fallout data reported by Martin (1965). Martin (1965) reported values from 1.9 to 11.1, with an average of 3.7 cm^2/g. For relative comparison purposes, Bloom's et al. (1974) factor of 5 cm^2/g can be converted to the unit-less factor by assuming a biomass of 289 g/m^2 (Martin et al. (1974)), resulting in an F value of 0.14.

Martin et al. (1974) uses parameters similar to those of Bloom et al. (1974). Several of the recommended parameters are: interception factor of 5 cm^2/g; weathering half-life of 30 days (based on nonvolatile particulates on shrubs); and a desert plant dilution growth rate of 36 $g(dry)/m^2$-year. A biomass value of 289 g/m^2 is referenced (Bamberg (1973)).

RADIONUCLIDE UPTAKE FROM SOIL BY PLANTS

The uptake of radioactive material from soil by plants is generally expressed as a discrimination factor pCi/g (dry weight) of plant per pCi/g (dry weight) of soil. Variations of this parameter include the discrimination factor between soil and the roots or the edible fruits of the plant. The discrimination factor is also given for uptake from hydroponic solutions. Table 2 summarizes the plant uptake of plutonium studies discussed below.

The depth basis of the soil concentration presents an inherent uncertainty in the discrimination factor. The majority of fallout plutonium is normally found in the top two-to-five cm of soil. Thus a 10-cm depth soil sample will only contain essentially one-half the concentration of plutonium as a 5-cm depth sample (i.e., the 10-cm sample is diluted with uncontaminated dirt). It appears that most discrimination factors are based on a 5-cm depth soil sample.

Several investigators have indicated an increase in the discrimination factor with time (Romney et al. (1970); Price (1973); Francis (1973)). The extent of and reasons for this increase are uncertain. It is generally related to either the chronological increase in depth penetration of plutonium in soil and/or the increased availability of plutonium with time.

The increased penetration is related to alternate freezing - thawing, and wetting - drying of the soil; earthworm activity; agriculture practices; possibly changing plutonium solubilization; and physical translocation downward through the soil by the root hair system of plants (Wildung and Garland (1974)). Wildung and Garland (1974) noted that plutonium from surface soil was translocated down to the roots of barley. This may have special health pathway implications for root crops directly consumed by man.

Chronological increases in the bio-availability of plutonium are related to the natural chelation of plutonium by decaying roots (Romney et al. (1970)). Au (1974) and Au and Becker (1975) indicate significant uptake of PuO_2 by soil microorganisms, specifically Aspergillus niger. Their experiments, conducted at several values of soil acidity, indicate that microorganisms may chronologically increase the bio-availability of PuO_2 microspheres.

TABLE 2. SUMMARY OF PLANT UPTAKE OF PLUTONIUM

Reference	Discrimination Factor (pCi/g dry plant: pCi/g dry soil)	Time of Measurement (Yrs)	Type Plant	Comments
Romney et al. (1970)	1.9×10^{-5}	1	Ladino Clover	Pu-239 from NTS soil
"	4.1×10^{-5}	2	" "	" " " "
"	4.4×10^{-5}	3	" "	" " " "
"	7.1×10^{-5}	4	" "	" " " "
"	14×10^{-5}	5	" "	" " " "
Garland et al. (1974)	4.4×10^{-5}	<1	Barley	$Pu(NO_3)_4$ 100 µCi/g in soil, toxic effects
"	15×10^{-5}	<1	Barley	$Pu(NO_3)_4$ 10 µCi/g in soil
"	1×10^{-7}	1/3	Barley seeds	
Johnson et al. (1972)	200 (roots)		Barley roots, activity may not have been taken up	
"	0.8 to 4×10^{-3} Aerial portion: root			Used PuO_2
"	2×10^{-6} to 10^{-3}			$Pu(NO_3)_4$
"	Av 6.4×10^{-4}			
Jacobson & Overstreet (1948)	10^{-4}	1/365	Barley	PuO_2^{2+}
"	2×10^{-5}	1/365	Barley	Pu^{4+}
"	4.5×10^{-6}	1/365	Barley	Pu^{3+}
"	0.4 (roots)		Barley	PuO_2^{2+}
"	0.25 (roots)		Barley	Pu^{4+}
"	0.2 (roots)		Barley	PuO_2^{2+}
Cline (1967)	Am-241 50xPu-239		Barley	
Cline (1967)	0.003 for Am			Alkaline Ephrata fine sandy loam
"	2×10^{-4}			Pu acid soil
Cline (1967)	1×10^{-4}			Pu alkaline soil
Neubold (1963)	Factor 4 increase	2	Ryegrass	Pu in acid soil
Nishita et al. (1965)	10^{-4}		Ladino clover	Pu Fallout
Rediske et al. (1955)	9×10^{-4}		Barley	Pu^{4+}

Barth (1975), Raabe (1973), Hakonson and Johnson (1973), and Patterson et al. (1974) report data inferring that plutonium-238 is more soluble than plutonium-239, and possibly more biologically available. Raabe et al. (1973) report that the dissolution rate for plutonium-238 dioxide monodispersed particles in an *in vitro* laboratory system was nearly two orders of magnitude greater than for plutonium-239. Hakonson and Johnson (1973) report changes in the plutonium-238 to plutonium-239 ratio for the Trinity Site, New Mexico. Twenty-three years after the nuclear detonation, the plutonium isotopic ratios varied from 0.05 for soil, 0.10 for plants, to 1.0 for mammals. Brown and McFarlane (1975) are conducting experiments with several plant species and soils to determine uptakes for plutonium-238 and plutonium-239.

Hanson (1975) notes that the increased availability of plutonium-238 may result from the chelating action resulting from more intimate contact of plant roots with the plutonium particles (plutonium-238 versus plutonium-239); transport of plutonium by individual cells; or a combination of such mechanisms by which plutonium-238 may be absorbed differently than plutonium-239. The higher specific activity of plutonium-238 versus plutonium-239 is also potentially related to possibly different isotopic effects.

Data indicating differences in the transfer or isotopic ratios for plutonium-238 and plutonium-239 should be critically evaluated. Plutonium-236, which is often used as an analytical tracer may contain plutonium-238 as a contaminant. This error can be corrected by analyzing tracer blanks. Furthermore, sources of purer plutonium-236 are now available. An additional problem results from the similarity of the americium-241 and plutonium-238 alpha energies, 5.49 and 5.50 MeV, respectively. Incomplete separation of americium in sample processing or delays in counting after sample processing (i.e., amercium-241 ingrowth from plutonium-241) can result in erroneously high indications of plutonium-238 content. Generally, these pitfalls are not present, but their potential must be recognized.

Plant uptake of plutonium from soil has been reviewed by Bloom et al. (1974), Hakonson (1975), Hanson (1975), Price (1973), and Francis (1973). The general consensus is that short-term uptake is minimal, but that increased chronological uptake due to natural chelation and other mechanisms presents an uncertain picture and some cause for concern.

Romney et al. (1970) studied the transfer of plutonium-239 from soil to plants for ladino clover. These crops were grown under glasshouse conditions on contaminated soil for five years. Total crop yields increased each year. The plant-to-soil discrimination factor for the first year was 1.9×10^{-5} pCi/g of dry plant per pCi/g of dry soil. The factor increased to 14×10^{-5} for the fifth year, for a five year average of 6.3×10^{-5}. The

soil was a one-to-one mixture of Yolo soil and soil from Area 11 of the Nevada Test Site contained in 60-liter containers (filled volume 50 liters), with a surface area of 0.12 m^2. The soil was uniformly contaminated with a plutonium-239 concentration of 1.62×10^{-5} dis/min-g. Romney et al. speculated that some of the yearly increase in plutonium uptake was related to increased development in the root system. They maintained the root system provided more intimate contact of the roots with plutonium. Additional studies showed increased plant uptake of plutonium from soils where DTPA chelating agent was added.

Garland et al. (1974) report results for barley and soybean plants grown in soils containing $Pu(NO_3)_4$. The split-root technique was used to study the uptake and distribution of plutonium in the plant tissue. The distribution of plutonium was determined in the tops and roots of soybeans (Glycine max) after 50 days of growth, and barley (Hordeim vulgare) after 27 days of growth. Slight increases in the total plant uptake were related to increasing the volume of soil in the test plots for both above-ground and root tissues of barley. But the height of the soil column appeared to be a more important variable. Since the plutonium was uniformly mixed in the soil column, increased uptake from a taller soil column probably relates to the increased contact between roots and soil.

Garland's et al. (1974) experiments were conducted with concentrations of 10 and 100 µCi/g of plutonium-239. But the elevated concentration of 100 µCi/g did not result in a marked increase in uptake versus the 10 µCi/g soil. The respective plant uptakes (dry weight, 60°C for 24 hours) for barley were 4.4×10^{-5} and 15.5×10^{-5}. The plants in the 100 µCi/g soil showed toxicity symptoms until the root systems were established in the nutrient solution below the soil. Plants grown in the two concentrations were indistinguishable at the time of harvest. If the observed toxicity is concentration dependent, Garland et al. (1974) indicate that the previously reported results of Wildung and Garland (1974), indicating an inverse relationship between uptake and soil concentration, may have been due to toxic effects on the roots.

Garland et al. (1974) reported that the distribution of plutonium activity in the plant roots of barley averaged 17.1 percent and 4.79 percent (percent of total plant activity) for two different plutonium activity soils (0.05 and 10 µg plutonium per µg soil). Therefore, the average root content was 10.95 percent, with the remaining plant activity in the above-ground parts of the plant (outer sheath, leaf blades and new growth). Both the barley and soybean plant studies indicated that plutonium, once in the plant, was rather mobile, with leaf tissue containing 5 to 10 times the plutonium activity of that in the stem tissue. After 100 days of growth, the barley seed had an activity corresponding to a concentration factor of less than 1×10^{-7} of the soil content.

Johnson et al. (1972) reported the results of an experiment to test the active transport of plutonium by plant roots. Barley plants were grown in a nutrient media containing soluble $^{239}Pu(NO_3)_4$. The barley plant roots were removed at various time intervals, washed and analyzed for plutonium-239 content. A tentative concentration factor from solution to roots of approximately 200 was observed. Although the roots were washed after removal from the nutrient media, it is possible that much of the plutonium was only associated with exterior surface contamination of the roots and was not assimilated by the roots.

Johnson et al. (1972) conducted another experiment where barley plants were allowed to mature and the root and aerial portions were removed and separately analyzed. The ratio or fraction of the concentration of plutonium in the aerial portion of the plants, as compared to the root portion of the plants, ranged from 0.8×10^{-3} to 4×10^{-3} (average of 2×10^{-3}) for plants grown in PuO_2 solutions. The ratio for plants grown in $Pu(NO_3)_4$ solutions ranged from 2×10^{-6} to 1×10^{-3}, with an average of 6.4×10^{-4}. These ratios are similar to those reported by Romney et al. (1970). This indicates that either the plutonium was not taken up into the roots, or there is a significant discrimination factor preventing the transfer of plutonium from roots to the above-ground parts of the plant.

Menzel (1965) reviewed the literature concerning the soil-plant relationships of radionuclides. This review was limited to experiments where the radioactivity was in a soluble form when added to the soil and where radionuclide concentrations were low enough so that there were no toxic effects. In summary, Menzel classed plutonium in the bottom of the lowest category (that is, less than 0.01 (ratio of dry weights of plant and soil activity)).

Francis (1973) reviewed the mobility of plutonium in soil and its uptake by plants. The following items are based on Francis' review:

1. Jacobson and Overstreet (1948) studied the translocation of plutonium in barley (one of the original plutonium plant uptake studies of barley plants in calcium-bentonite clay suspensions). Over a 24-hour period, the fractional translocations to leaves, were 10^{-4} for PuO_2^{2+}, 2×10^{-5} for Pu^{4+}, and 4.5×10^{-6} for Pu^{4+}. The respective values for the roots were 0.4, 0.25, and 0.2.

   Rediske et al. (1955) noted the discrimination factor (ratio of dry weight of aerial portion of plants to soil) increased from 10^{-4} to 10^{-3}, with pH changes of 7 to 4.

2. Wilson and Cline (1966) studied the uptake of plutonium-239 by barley from three soils. The soils were Ephrata fine sandy loam, a slightly alkaline soil; Milville silt loam, a calcareous soil; and Cinabar silt loam, a moderately acid forest soil. Plutonium uptake from the acid soil was more than three times greater than from the calcareous soil. A 0.1N nitric acid solution removed 0.64 percent of the plutonium, approximately one-thousand times more than that taken up by barley. This shows that common soil extracting methods do not provide a reliable indication of potential plant uptake.

3. Cline (1967) reported that the uptake of americium-241 into foliar portions of barley was fifty times that of plutonium-239. The barley was grown in Hoagland's nutrient solution.

4. Unpublished work of Buckholz et al. does not show a chronological increase in the discrimination factor for alfalfa after four years of growth. The study was conducted in a plutonium contaminated soil associated with the 1966 Palomares Spain accident. This is at variance with the results of Romney et al. (1970).

Price (1973) reviewed several studies concerning plant and animal uptake of plutonium. The following studies were not reported by Francis (1973):

1. Nishita et al. (1965) studied the uptake of fallout plutonium in ladino clover (Trifolium respens L.). The discrimination factor was 10^{-4} (μCi/g plant per μCi/g soil, probably dry weights).

2. Rediske et al. (1955) noted that Pu^{4+} becomes associated with root surfaces exposed to culture solutions. The quality of sorption to root surfaces is linear with respect to concentration of the solution, whereas, the leaf concentrations had a curvilinear relationship. The uptake into shoot tissues of tumbleweed from solution cultures was slightly less than for beans, barley, or tomatoes. The discrimination factor for barley was 9×10^{-4}, based on the Neubauer test. This was considered to be an overestimate for what would be expected under field conditions.

3. Cummings and Bankert (1971) used culture pots for plutonium-238 uptake studies for nine soils. The results for plutonium-238 were lower than those for cerium-144 and promethium-147. The fractional uptake (total activity in plants divided by total soil activity) for plutonium varied from 7 to 280×10^{-8}.

4. Cline (1967) reports discrimination factors of 0.003 for americium-241 from alkaline (Ephrata lime sandy loam) and acid (Cinabar silt loam) soils. The plutonium factors were 2×10^{-4} (acid soil) and 1×10^{-4} (alkaline soil).

5. Neubold (1963) reported that although plutonium uptake by perennial ryegrass (Folium perenne L.) was low, it did increase over a 2-year study span, for several different soils. There was a 4-fold increase for an acid soil.

Price (1973) indicates the following ranking for decreasing uptake by plants from soil: curium, americium, and plutonium. Neptunium uptake probably resembles that of plutonium.

Hakonson (1975) reviewed pathways for plutonium into terrestrial plants and animals. Several investigators have noted higher plutonium concentrations in native grasses than for forbs, shrubs, or trees (e.g., Hakonson and Johnson (1973) and Whicker et al. (1973)). This may be related to the morphological structures of the plants and their ability to intercept and entrap airborne material. Russell (1966) has noted that the heads of grains serve as an excellent trapping device for deposited material. On the other hand, the physical structure of root systems of grasses and their position within the soil/plutonium profile may be favorable for root uptake of plutonium by grasses.

Bloom et al. (1974) present an environmental transport model with associated parameters. A soil to plant discrimination factor of 0.313 (pCi/g of dry vegetation per pCi/g of wet soil) is recommended. This factor is based on the indications of increases of plant uptake with time (e.g., Martin's et al. (1974) estimates from data from Romney et al. (1974)). The Romney et al. (1974) values are based on results from the Nevada Test Site, 20 years after deposition. Martin et al. (1974) further justified the value of 0.313 by estimating the uptake at 20 years from Romney's et al. (1970) data. In essence, Martin et al. (1974) exponentially extrapolated Romney's et al. (1970) 5-year study to 20 years.

Romney et al. (1974 and 1975) emphasize that the high discrimination factors are not solely related to root uptake, rather, they are a result of deposition with limited root uptake. Romney et al. (1975) estimate the root uptake to be 10^{-3} to 10^{-4}. Thus, it appears Bloom's et al. (1974) and Martin's et al. (1974) assumptions are in error.

COMBINATION OF DEPOSITION AND PLANT UPTAKE

Romney et al. (1974 and 1975) reports data on the total plutonium and americium-241 concentrations in vegetation for several areas of the Nevada Test Site. These data are compared on a pCi/g (dry and ashed) basis to soil concentrations. The vegetation results are based on a sample of foliage, and exclude the root mat. Romney et al. (1975) estimated that for Area 13 only 1/1600 of the plutonium-239 inventory was in the vegetation. The ratio of foliage to soil values averaged about 0.08, and most of the values (several hundred) fell within 0.02 to 0.16.

Romney et al. (1974) reported values for the total amount of plutonium-239 on foliage versus the soil concentration. The preliminary results for vegetation were based on the ashed weight. The average values (pCi/g ashed vegetation per pCi/g dry soil) were 1.0, 1.7, and 5.1 for Atriplex canescens, Atriplex confertifolia, and Eurotia lanata, respectively. The average value was 2.24. Romney's plant-to-soil ratios are apparently all based on the plutonium concentration in the top 5 cm of soil.

Colorado State University (1973) reported data on the plutonium-239 inventory for the Rocky Flats, Colorado, area. The data is based on plant distributions from the Pawnee National Grassland. The soil accounted for 99.464 percent of the plutonium-239 in the top 2 cm of soil. Standing vegetation accounted for 0.058 percent, litter for 0.180 percent, and roots (surface to two cm depth) accounted for 0.298 percent of the total activity on the test plot. Considering the litter as part of the standing vegetation, the foliage would then account for 0.238 percent of the total activity, comparable to the root content of 0.298 percent. The total plant content would be 0.536 percent.

Whicker et al. (1973) reported plutonium concentrations for various terrestrial ecosystems in the Rocky Flats environs. In the top 3 cm of soil, fifty-nine percent of the plutonium-239 was found in the soil fraction of less than 0.5 cm in diameter. Additionally, 39 percent of the soil activity was found to be below the 3 cm depth. Two-tenths percent was associated with the surface litter and detritus, 1.3 percent with roots, and 0.06 percent with standing vegetation. Considering the litter as part of the standing vegetation, the foliage would then account for 0.26 percent of the total activity, compared to the root content of 1.3 percent. The total plant content is 1.56 percent, corresponding to a concentration factor of 0.0156, due to both

deposition and soil uptake. The root, foliage, and soil data from Whicker (1973) are summarized in Table 3.

TABLE 3. PLUTONIUM IN VEGETATION AND SOIL

(Whicker (1973))

Plant	Biomass Dry Weight (g/m^2)	Average Conc. In Roots (dpm/g)	Average Conc. In Standing Plant (dpm/g)	Standing Veget. Conc: Soil conc.*
Western Wheatgrass	32	247	30	0.00125
Cheatgrass	10	294	112	0.0467
Prickly lettuce	2	157	13	0.00542
Salsify	9	13	13	0.00542
			Average	0.0015±0.02

* Vegetation concentration in dpm/g divided by average soil concentration of 2397 dpm/g. Based on soil sample of 0 to 3 cm depth and particles less than 5 mm. Dry Weights.

Schultz et al. (1974) report a proposed study of plant uptake of plutonium and americium. The study will include several soils and several chemical forms of the elements. Results have not yet been published.

REDISTRIBUTION OF ACTIVITY WITHIN PLANTS

There appears to be very little absorption and redistribution of deposited plutonium within plants. There is, however, only a limited amount of published information.

Russell (1965) reviewed several studies concerning interception and retention of airborne material. He concluded that the absorption of deposited material was of limited importance compared to the retention of activity on foliar surfaces.

Aarkrog (1975) studied the uptake of deposited fission products on wheat and barley crops. Radionuclides such as strontium-90, ruthenium-103, and cerium-144 were generally immobile. Whereas, zinc-65, iron-55, cesium-137, cobalt-60, and manganese-54 were more readily translocated to the seeds.

Levin et al. (1970) estimate that only 0.1 percent of the radioactivity in plant edibles comes from the leaves (for relatively immobile elements). Essentially all of the activity in the plant edibles is related to root uptake. The parameters listed in Appendix A (Levin et al. (1970)) are for the interoceanic canal project and are assumed to relate to fruits, nuts, etc., -- not to leafy edibles.

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Appendix A

Parameters for Atlantic-Pacific Interoceanic Canal Model (LEVIN et al., 1970)

Parameter	Parameter Definition	Values Used	Remarks	References
F_A	Average fallout concentration on a watershed, $\mu Ci/cm^2$	-	These values are classified and thus are not available	-
S_{01}	Initial specific activity of the radionuclide in the fallout, $\mu Ci/g$ element	-	These values are classified and thus are not available	-
f_{43}	Fraction of the element in plant edibles which comes from leaves (dimensionless)	0.05 (P&C)[a]	For mobile elements H, P, I, and C	Martin (1969)
		0.001 (P&C)	For all other elements than H,P, I, and C	Martin (1969)
f_{42}	Fraction of the element in plant edibles which comes from the soil (dimensionless)	1.0 (P&C)	For all elements except carbon	Kazmaier (1969)
		0.001 (P&C)	For carbon	Kazmaier (1969)
f_r	Ratio of runoff water to total rain water (dimensionless)	0.9 (P&C)	Infiltration is 10 percent of rainfall reaching ground surface	Charnell et al (1969)
F	Ratio of the amount of radionuclide dissolved in surface water to the total amount present on the soil surface (dimensionless)	4.0×10^{-5} (P&C)	For all elements except hydrogen; calculated from reference data[b]	Charnell et al (1969)
		0.3 (P&C)	For hydrogen; calculated from reference data[b]	Charnell et al (1969)
r	Average rainfall rate, cm/day	0.636 cm/day (P) 0.596 cm/day (C)	Estimated from mass rainfall curves Estimated from rainfall curves	Charnell et al (1969) Charnell et al (1969)
r_u	Unit rain, cm	2.5 cm (P&C)	Defined by reference[c]	Charnell et al (1969)
W_4	Dry biomass of plant edibles, g dry weight/cm^2	0.054 g/cm^2 (P&C)	Calculated from reference data[d]	Transeau (1926)
X_{W4}	Fraction of water in plant edibles, g water/ g fresh weight	0.70 g/g (P&C)	Estimated average water content from reference data	Wu Leung and Flores (1961)

Appendix A (continued)

Parameter	Parameter Definition	Values Used	Remarks	References
ρ_w	Density of water, g/cm^3	1.0 g/cm^3 (P&C)	Listed in reference	Weast and Selvy (1967)
λ_{B6}	Biological elimination rate coefficient of element from freshwater fish, day^{-1}	0.055 day^{-1} (P&C)	Geometric mean calculated from reference data by method outlined by Bloom et al (1970)	Templeton et al (1969) Brungs (1967) Boroughs et al (1956) Polikarnpov (1966 a&b) Kevern (1966) Wiser and Nelson (1964) Friend et al (1965)
λ_{B7}	Biological elimination rate coefficient of element from animals, day^{-1}	0.1 day^{-1} (P&C)	Geometric mean calculated from reference data	Golley et al (1969)
λ_{B9}	Biological elimination rate coefficient of element from marine fish, day^{-1}	0.02 day^{-1} (P&C)	Estimated from turnover rates for anchoveta	Lowman et al (1970)
λ_g	Growth-rate coefficient for plant edibles, day^{-1} $\lambda_g = (\frac{d C_4}{dt})/C_4$	0.05 day^{-1} (P&C)	Arithmetic mean calculated from reference data by method outlined by Bloom et al (1970)	Malavolta et al (1962) Transeau (1926) Jacob and von Uexkull (1963)
k_w	Weathering elimination rate coefficient from plant leaves, day^{-1}	0.05 day^{-1} (P&C)	Calculated from reference data[f]	Martin (1965)
f_I	Fraction of fallout intercepted by plant leaves (dimensionless)	0.25 (P&C)	Highest value selected from reference data[g]	Nishita et al (1965) Middleton (1960)
W_3	Biomass of plant leaves, g dry weight/cm^2	0.35 g/cm^2 (P&C)	Calculated from reference data[h]	Transeau (1926)
F_{AM}	Average fallout concentration on the marine fallout area, $\mu Ci/cm^2$	-	These values are classified and thus are not available	-
Q	Total amount of radionuclide initially present in the canal channel and rubble, μCi	-	These values are classified and thus are not available	-
F_c	Ratio of the amount of radionuclide dissolved in the canal water to the total amount present in the canal channel and rubble (dimensionless)	4.0×10^{-3} (P&C)	For all elements except hydrogen[i]	Essington (1969)
		0.3 (P&C)	For hydrogen	Essington (1969)

Appendix A (continued)

Parameter	Parameter Definition	Values Used	Remarks	References
B_c	Net flow rate of water through the canal channel, cm^3/day	2.83×10^{14} cm^3/day (P)	Estimated from reference data	Harleman (1967)
		3.62×10^{11} cm^3/day (C)	Estimated from reference data	Harleman (1967)
H	Horizontal extent of the marine fallout field in the direction perpendicular to the current, cm	1.85×10^7 cm (P-Pacific side)	Estimated from reference data(j)	Ferber (1968)
		1.20×10^7 cm (P-Atlantic side)	Estimated from reference data(j)	Ferber (1968)
		1.11×10^7 cm (C-Pacific side)	Estimated from reference data(j)	Ferber (1968)
Z	Horizontal extent of the marine fallour field in the direction parallel to the current, cm	1.11×10^7 cm (P-Pacific side)	Estimated from reference data(j)	Ferber (1968)
		7.4×10^6 cm (P-Atlantic side)	Estimated from reference data(j)	Ferber (1968)
		9.26×10^6cm (C-Pacific side)	Estimated from reference data(j)	Ferber (1968)
K	Turbulent diffusivity in the vertical direction, cm^2/day	1.0×10^6 cm^2/day (P&C)	Estimated from reference data(k)	Pritchard et al (1966)
V_c	Volume of water in the canal channel, cm^3	2.52×10^{15} cm^3 (P)	Estimated from reference data	Harleman (1967)
		2.19×10^{15} cm^3 (C)	Estimated from reference data	Harleman (1967)
V_o	Speed of ocean current, cm/day	1.0×10^6 cm/day (P)	Estimated from reference data(k)	Lowman et al (1970)
		3.0×10^6 cm/day (C)	Estimated from reference data(l)	Lowman et al (1970)

(a) P designates value used for Panama (Route 17). C designates value used for Colombia (Route 25).

(b) The quantity F_w was calculated as follows:

$$F_w = \frac{1}{\left[1 + \left(\frac{1-\sigma}{\sigma} K_D\right)\right]} ,$$

where σ is the fractional soil porosity, 0.3

and K_D is the distribution coefficient of the rainwater between the soil surface and surface water, 1 for hydrogen, and 1.0×10^4 for all other elements.

(c) If the interflow layer has a thickness of 7.5 cm (Odum, 1967) and a porosity of 33 percent, then the amount of rain required to saturate this interflow layer is the unit rain (2.5 cm).

Footnotes for Appendix A (continued)

(d) The quantity W_4 was calculated as follows:

$$W_4 = \left[\frac{\text{dry weight plant edible}}{\text{plant}}\right] \times \left[\frac{\text{No. plants}}{\text{cm}^2}\right]$$

$$W_4 = \left[\frac{216\text{g plant edible}}{\text{plant}}\right] \times \left[\frac{1.0 \times 10^4 \text{ plants}}{\text{acre}} \times \frac{2.471 \times 10^{-6} \text{ acres}}{\text{cm}^2}\right]$$

(e) Reference data described the growth of tropical plants. Pineapple, sugar cane, rice, and bananas were some of the foodstuffs for which growth rate data were reported.

(f) The quantity k_w was calculated as follows: $k_w = \frac{0.693}{T_e}$

where T_e is the environmental half-life of a radionuclide on leaves.

T_e was assumed to equal to 14 days for all radionuclides on fallout-contaminated plants in humid regions.

(g) The relative percentage of fallout intercepted by plants in the environs at NTS (Nishita et al, 1965), from 8 percent to 15 percent at the Maralinga Test Site for close-in and far-out fallout, respectively, (Nishita et al 1965), and up to 25 percent retention near NTS (Middleton, 1960).

(h) The quantity W_3 was calculated as follows:

$$W_3 = \left(\frac{\text{dry weight plant leaves}}{\text{plant}}\right) \times \left(\frac{\text{no. plants}}{\text{cm}^2}\right)$$

$$W_3 = \left(\frac{140 \text{ g leaves}}{\text{plant}}\right) \times \left(\frac{1.0 \times 10^4 \text{ plants}}{\text{acre}} \times \frac{2.47 \times 10^{-6} \text{ acre}}{\text{cm}^2}\right)$$

(i) The quantity F_c was calculated for all elements except hydrogen as follows: $F_c = 100\ F_w$.

(j) This quantity is dependent upon the orientation of the fallout patterns over the marine fallout area. As no fallout is expected over the Atlantic side of Route 25, no values are listed for the Atlantic side of Route 25.

(k) This quantity is the value used for both the Atlantic and Pacific sides of Route 17.

(l) This quantity is the value used for the Pacific side of Route 17.